

TECHNICAL REPORT RD-AS-87-14

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GADOLINIUM SCANDIUM GALLIUM GARNET (GSGG) AS A SOLID-STATE LASER HOST

H. Lee Pratt Advanced Sensors Directorate Research, Development, and Engineering Center

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Redstone Arsenal, Alabama 35898-5000

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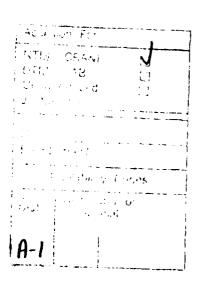
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## I. INTRODUCTION

This report provides a discussion on the limitations of Nd:YAG lasers and the possibility of using certain other garnet materials for replacement. It also addresses the solid-state laser host material Gadolinium Scandium Gallium Garnet (GSGG) and its application in two separate types of lasers. First, GSGG is treated as the host material for chromium-doped lasers which are tunable in the near-infrared spectral region. Second, neodymium-doped GSGG, with and without chromium as a sensitizing codopant, is considered as a potential direct replacement for Nd:YAG to improve performance.

### II. BACKGROUND

The current myriad of laser systems owe their beginning to work in a solid-state crystal of chromium-doped sapphire (Al<sub>2</sub>O<sub>3</sub>) material, better known as ruby. Ruby has remained a mildly important material for a number of years for a few special applications. Most notable, ruby was used by the military for single-pulse rangefinder systems. It is still being used in armored M-60 tanks in the US and in a number of systems in Europe. However, its very low electrical efficiency and other undesirable characteristics have led to its replacement by neodymium-doped yttrium aluminum garnet (Nd:YAG) or other materials for most applications.

In the years after the invention of the ruby laser, in 1961, there was intensive research in other dielectric solid-state lasers, both crystalline and glass. Many of the subsequently discovered solid-state crystalline laser materials, such as YAG, were also a form of aluminum oxide but were in the garnet family. In addition, trivalent rare earth ions (such as Cr<sup>3+</sup>, Nd<sup>3+</sup>, Tm<sup>3+</sup>, Er<sup>3+</sup>, Ho<sup>3+</sup>, Yb<sup>3+</sup>, Pr<sup>3+</sup>, Gd<sup>3+</sup>, and Eu<sup>3+</sup>) were often employed as the lasing medium or as a codoped sensitizer to aid the lasing medium for improved efficiency [1, p. 33]. Several hundred different laser hosts were identified and characterized, but most were forgotten after a few years of research when only mediocre performance levels were obtained.

Nd:YAG and Nd:Glass lasers emerged as the predominant solid-state laser materials for worldwide applications. Nd:YAG is usually employed where moderate repetition rates and good beam quality are required. In contrast, glass lasers may produce higher energy levels, but at lower repetition rates and usually poorer beam quality. However, at least three fundamental reasons continue to spur research toward replacement materials for Nd:YAG and glass: (1) the desire for other wavelengths for applications such as eye safety, secondary reactions, scientific research, etc.; (2) the desire for tunable lasers for the above and other applications; and (3) improved efficiency and other laser performance factors.

Several important developments in solid-state lasers have occurred in recent years, including tunable color-center lasers, the vibronic lasers, some tunable divalent lasers, and a new family of garnet materials. Perhaps the most promising of the new garnets is GSGG. Also of interest is the fact that GSGG as a laser host is being intensely investigated with the doping impurities  $Cr^{3+}$  and  $Nd^{3+}$ , the two materials which have dominated solid-state lasers from their very beginning.

According to Krupke et al [2, p. 102], the Soviets first utilized GSGG for laser purposes in 1976 when doped with Nd<sup>3+</sup>. The University of Hamburg (West Germany) and the Lebedev Institute (Moscow), in a cooperative effort, first reported GSGG as a tunable source when doped with Cr<sup>3+</sup> and pumped by dye, krypton, and argon lasers [3 and 4]. In 1983, the Soviets reported flash-lamp pumped operation of the tunable chromium laser, and they continue to publish important findings from the General Physics Institute and Lebedev Physics Institute in Moscow. Other results have been released separately by the University of Hamburg in West Germany. Published reports have been available from the United Kingdom and from a number of US sources in the last three years. In addition, a number of laser manufacturers are conducting research which may soon lead to commercially available systems.

### III. DISCUSSION

### A. Limitations of Nd:YAG

Although widely used in many commercial and military applications, Nd:YAG lasers exhibit a number of limitations for which GSGG shows a potential improvement. Flashlamp pumped Nd:YAG lasers typically have an electrical efficiency of only 1 to 3 percent in Q-switched operation. Although this efficiency is well above that of ruby and most other solid-state lasers, it is low enough that special design considerations must be enforced. A cooling system is necessary for all but very modest, low power and low repetition rate operation. Conductive cooling through the mechanical structure may be sufficient to remove heat from the flashlamp and laser rod of a smaller system. However, larger systems require a recirculating fluid (liquid or gas) for effective heat removal.

The low electrical efficiency also requires a larger power supply, storage capacitor, pulse forming network, and flashlamp than would be required for a more efficient system. In addition, if the laser is battery powered for portable applications, the laser efficiency contributes directly to the battery size, weight, and run time.

Note that only a modest increase in overall system efficiency, perhaps 2 to 3 percent, would double that obtainable from Nd:YAG. The improved laser could obtain the same output levels, by pumping half as hard, using a power supply with half the capacity, and using batteries approximately half as large. Alternatively, the same batteries, used with the Nd:YAG laser, would give about twice the run time with the more efficient laser. Since this laser would be pumped at half the previous level, the cooling system would only need to be half as efficient and a smaller size would be possible. GSGG shows promise for this improvement in efficiency.

Nd:YAG laser rods are also difficult to fabricate. Nd:YAG crystals are grown in boules typically up to 2 to 3 inches in diameter and up to 4 to 5 inches in length. The Czochralski method is used, whereby a high-quality seed crystal is attached to a revolving stem and lowered to the surface of a liquid melt in a high-temperature furnace. The solid crystal is slowly pulled from the liquid melt which, due to the extremely high temperature, must be contained in a crucible of platinum and iridium. The furnace may be heated by RF or resistance means.

One of the reasons that Nd:YAG is difficult to grow is the very low distribution coefficient for  $Nd^{3+}$  in YAG. The distribution coefficient is the ratio of the concentration in the solid to the concentration in the melt, which for  $Nd^{3+}$  in YAG is only 0.18 [5, p. 67]. This means the liquid melt must be rich in  $Nd^{3+}$ , and the concentration will change as the boule is pulled so that the top end of the boule has a lower concentration of  $Nd^{3+}$  than the bottom end. A variation in concentration impedes proper laser operation. In addition, the boule must be pulled very slowly, over a period of time up to several weeks, leading to high fabrication costs and electrical power costs. Again, there is potential for GSGG as a crystal that is easier to grow with distribution coefficients for  $Cr^{3+}$  and  $Nd^{3+}$  that are closer to unity.

#### B. GSGG and Related Garnets

GSGG has the nominal chemical constituency Gd<sub>3</sub>Sc<sub>2</sub>Ga<sub>3</sub>O<sub>12</sub>, but other composition ratios are also possible. Sometimes the terminology Gd<sub>3</sub>(Sc,Ga)<sub>2</sub>Ga<sub>3</sub>O<sub>12</sub>, Gd<sub>3</sub>(Sc,Ga)<sub>5</sub>O<sub>12</sub>, or similar nomenclature is used to indicate slight differences in stoichiometry. GSGG can be grown by the Czochralski method in large diameters (5 cm or more) of high optical quality without an index of refraction distortion (core) occurring along the boule axis [2, p. 102]. In addition the distribution coefficient for Cr<sup>3+</sup> in GSGG is essentially unity, so uniform doping occurs without a variation in concentration along the rod axis. The distribution coefficient for Nd<sup>3+</sup> in GSGG is about 0.65 [2, p. 102], which is nearly four times higher than for Nd<sup>3+</sup> in YAG. Therefore, Nd:GSGG and Cr,Nd:GSGG can be grown easier and faster than Nd:YAG.

On the negative side, GSGG crystals sometime exhibit formation of color centers which are believed to be associated with a gallium deficiency [6, p. 198]. In addition GSGG is a complex formation, particularly when codoped with  $\mathrm{Nd}^{3+}$  and  $\mathrm{Cr}^{3+}$ . A fair statement is that additional "fine-tuning" of the optimum recipe for best laser performance is probably necessary.

Although GSGG is perhaps the most promising of the new laser garnets, others are also being investigated. These include  $Gd_3Sc_2Al_3O_{12}$  or GSAG,  $Gd_3Ga_5O_{12}$  or GGG,  $(La,Lu)_3(Lu,Ga)_2Ga_3O_{12}$  or LLGG,  $Y_3Sc_2Ga_3O_{12}$  or YSGG,  $Y_3Ga_3O_{12}$  or YGG, and perhaps others [7, p. 21] and [6, pp. 196 and 197]. When doped with  $Cr^{3+}$ , all of these other garnets can be made to lase between 730 and 850 nm, as shown in Table 1.

TABLE 1. New Garnet Laser Materials

Common Name	Chemical Formula	Wavelength, nm	
Cr:GSGG	Cr <sup>3+</sup> :Gd <sub>3</sub> (Sc,Ga) <sub>2</sub> Ga <sub>3</sub> O <sub>12</sub>		
Nd:GSGG	$Nd^{3+}:Gd_3(Sc,Ga)_2Ga_3O_{12}$	1061	
Cr, Nd: GSGG	$Cr^{3+}$ , $Nd^{3+}$ : $Gd_3(Sc, Ga)_2Ga_3O_{12}$	1061	
Cr:GSAG	Cr <sup>3+</sup> :Gd <sub>3</sub> Sc <sub>2</sub> Al <sub>2</sub> O <sub>12</sub>	735-820	
Cr:GGG	Cr <sup>3+</sup> :Gd <sub>3</sub> Ga <sub>5</sub> O <sub>12</sub>	745	
Cr:LLGG	Cr <sup>3+</sup> :(La,Lu)3(Lu,Ga)2Ga3012	850	
Cr:YSGG	Cr <sup>3+</sup> : Y <sub>3</sub> Sc <sub>2</sub> Ga <sub>3</sub> O <sub>12</sub>	750	
Cr:YGG	Cr <sup>3+</sup> :Y <sub>3</sub> Ga <sub>3</sub> O <sub>12</sub>	730	

Sources: Walling, [6, pp. 196 and 197], Beimowski et al, [8, p. 235], and Pruss et al, [9, p. 358].

### IV. DOPED GSGG LASERS

#### A. Tunable Chromium

This paragraph reviews the properties of Cr<sup>3+</sup>:GSGG as a tunable laser. Struve et al [3, pp. 235 and 236] first reported laser action and broad band fluorescence when pumping a small crystal with a pulsed Rhodamin 6G dye laser at 590 nm. Although a broad fluorescence spectrum occurred, the laser ouput was limited to about 790 nm due to the wavelength-dependent reflectivities of the mirror coatings. Within a year the same team reported room-temperature tunable cw laser action using krypton, argon, and Rhodamin 6G dye lasers as pump sources [4, pp. 117-120]. Flashlamp pumped operation was reported soon thereafter. CW pumping via a krypton laser was commercially available by 1984 [10, pp. 79-81].

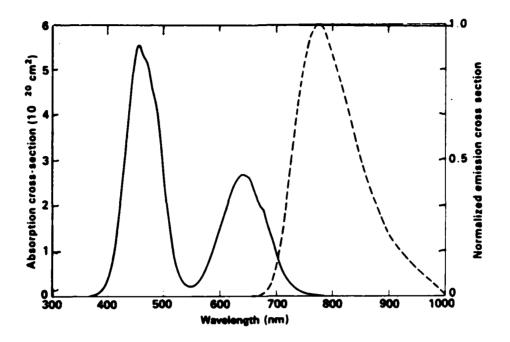
# 1. Properties of Cr3+:GSGG Crystals

Cr<sup>3+</sup>:GSGG boules are grown by the Czochralski technique in iridium crucibles at 1750 °C [3, p. 235 and 4, pp. 117 and 118]. The crystals exhibit many of the favorable properties of other garnets: high mechanical and chemical stability, good heat conductivity, good optical quality, and somewhat standard technology for crystal growth. Krupke et al [2, pp. 107-111] have compiled an extensive summary of the bulk optical properties of GSGG. These include the refractive indices at several wavelengths (typically between 1.9 and 2 through the visible and near infrared spectrum), the thermal dependence of the refractive index, and the piezo-optical and elasto-optical constants. Thermomechanical properties such as tensile strength, fracture toughness, thermal conductivity, thermal expansion, and thermal stress resistance are also reported. It is concluded that GSGG compares favorably with Nd:YAG.

In  $Cr^{3+}$ :GSGG the crystal is typically doped with 0.3-3 X  $10^{20}$  ions/cm<sup>3</sup> of  $Cr^{3+}$ . Since the previously mentioned distribution coefficient for  $Cr^{3+}$  in GSGG is essentially unity, it is easy to vary the  $Cr^{3+}$  concentration. These concentration levels correspond to less than 1 percent by atomic weight.

As in other garnets, GSGG is a cubic crystal, but somewhat distorted from perfect symmetry due to the mix of atomic sizes and the doping impurity [4, pp. 117 and 118]. The chromium ions result in a strong absorption of pump light at each end of the visible spectrum, as shown by the solid line in Figure 1. Laser pumping is therefore possible either with a bluegreen source in the first absorption band or a yellow-red source in the second band. As mentioned earlier, Struve et al [3 and 4] demonstrated pumping by an argon laser (approximately 500 nm wavelength) in the lower absorption band and pumping by Rhodamin dye (approximately 600 nm) and krypton (647 nm) lasers in the upper band. Another absorption band exists in the ultraviolet spectrum at 300 nm and shorter wavelengths.

The dashed line in Figure 1 shows the normalized emission cross section at an ambient temperature of 295 °K. This emission cross section indicates the spectral region for which laser action may be tuned. Peak emission is in the vicinity of 770 nm, and the half value points are near 740 and 840 nm, which define the region over which laser action can be most easily tuned.



Source: From page 107 of Reference 2.

Figure 1. Absorption cross section (---) and normalized emission cross section (---) of Cr<sup>3+</sup> in GSGG.

Krupke et al [2, p. 107] measured a fluorescent lifetime of l12  $\pm$  5  $\mu s$ . This is in good agreement with the 120  $\mu s$  value reported by Struve et al [4, p. 118]. This fluorescent storage lifetime is somewhat comparable to that of Nd:YAG at 230  $\mu s$  [1, p. 54], which means the Cr $^{3+}$ :GSGG laser should be amenable to pumping by flashlamp pulses and subsequent Q-switching. However, optimum pumping would require a flashlamp pulse approximately half as long as used with Nd:YAG lasers.

# 2. Lasing Transitions

The lasing transitions important in Cr3+:GSGG, as in other tunable solid-state lasers, are known as "vibronic" transitions. In a vibronic transition the active species (chromium in this case) changes both vibrational and electronic states. The term "vibronic" is a hybrid word coined to indicate that two kinds of states are involved [10, p. 77]. In addition to the new tunable garnets, other vibronic lasers include alexandrite, or Cr3+:BeAl<sub>2</sub>O<sub>4</sub>, emerald, titanium-doped sapphire, and other lasers which are not part of this study.

Due to a low crystal field at the Cr site, the chromium ion is not "tightly bound" and broad bands of energy levels exist. The major transition is from the  $^4\mathrm{T}_2$  excited state (band) to the  $^4\mathrm{A}_2$  state of Cr $^{3+}$  in GSGG. The energy gap between the  $^2\mathrm{E}$  and  $^4\mathrm{T}_2$  levels is very small and energy is transferred from the  $^2\mathrm{E}$  to the  $^4\mathrm{T}_2$  state to further enhance the lasing transition.

The net result is a broad band tunable laser operating effectively in a 4 level arrangement which is not possible for chromium in ruby. A population inversion can be continuously maintained with a cw pump source, resulting in cw laser operation. Alternatively, the fluorescence lifetime in excess of 100 µs also makes pulsed operation possible.

## 3. Experimental Results

The first experiments by Struve et al [3, p. 236] used a 10  $\mu s$  pulsed dye laser. Only about a 1 percent slope efficiency was obtained. In the succeeding report [4, pp. 118 and 119] cw slope efficiencies up to 11 percent were obtained with a krypton laser as pump source, and 6.5 percent slope efficiency was measured with an argon laser pump.

More recent data exists which have been reviewed every year or so in major international conferences dedicated to the subject of tunable solid-state lasers. The most recent conference was held in June, 1986 at Zigzag, Oregon and sponsored by the Optical Society of America [11, p. 54]. At that time new developments in Cr<sup>3+</sup>:GSGG lasers included the following:

- a. Caird [7, p. 20] reported on the special attributes of  ${\rm Cr}^{3+}$  as the activating media in solid-state lasers, due to its trivalent  ${\rm d}^3$  electron configuration. Chromium has large absorption bands, good chemical stability and other features which make it an ideal choice.
- b. Armagan and Di Bartolo [12, p. 35] have studied the effects of temperature on energy transfer in GSGG. The fluorescence lifetime of Cr was found to be exponential in the temperature range from 77 to 660 °K. At the high temperature the lifetime decreases to only 4.9  $\mu s$ . Below 400 °K the lifetime is longer than 100  $\mu s$ , which is acceptable for flashlamp pumping.
- c. Andrews [13, p. 44] described an energy level model which predicts the absorption and fluorescence spectra as measured in Cr:GSGG.
- d. Fuhrberg et al [14, p. 113] from the West German company, Spindler & Hoyer, reported on their work with krypton laser pumps. Output powers as high as 60 mW (cw) were reported at the peak wavelength for operation near single mode. Output levels dropped to 10 mW near 765 nm and (on the high end of the spectrum) to less than 30 mW at 825 nm. A birefringent filter was used as the tuning element. Spindler & Hoyer commercially sell multimode 200 mW Cr:GSGG lasers [15, p. 72].
- e. Drube et al [16, p. 118] reported flashlamp pumped output energies up to 10 mJ, but only at repetition rates of 0.3 Hz. At rates of 1 and 2 Hz the output level dropped significantly. This effect is due to ultraviolet and blue light induced color center absorption, which overlaps and interferes with the desired laser transitions. The transient color centers decay at times on the order of 10 ms up to seconds, depending upon the raw crystal material, purity, and growth procedure. Blocking filters were used to minimize flashlamp light from the ultraviolet and blue end of the spectrum. Much better performance (up to 200 mJ) was obtained with Cr<sup>3+</sup>:GSAG, which did not exhibit the transient color centers.

f. Payne and Evans [17, p. 126] also reported on flashlamp pumped operation and filtering of the shorter wavelength pump light. By investigating several techniques of spectral blocking, outputs up to 60 mJ and higher were achieved. However, color centers soon developed after a number of shots and output levels dropped. Payne and Evans attributed the color centers as possibly due to oxygen vacancies in the GSGG host. Absorption spectra of undoped GSGG confirmed that the color centers were not associated only with the presence of chromium.

It is concluded from the 1986 conference on Tunable Solid State Lasers that Cr:GSGG may prove to be a useful source for cw tunable laser radiation in the 740-840 nm band. Small systems are not available because of the requirement for krypton or argon ion laser pumps.

As a compact pulsed laser source, however, Cr:GSGG does not currently look very promising. Although the fluorescence lifetime is suitably long, the formation of color centers from ultraviolet and blue light emission in flashlamps seriously limits laser performance.

No reference has been found to any group investigating laser diode pumping of GSGG. Such a technique may hold some promise in the future for compact systems.

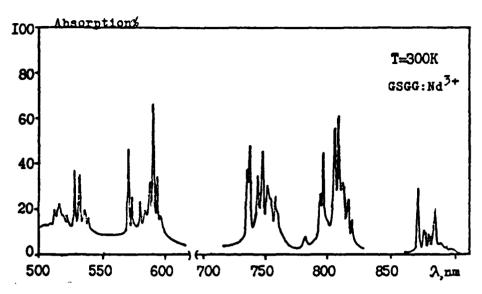
### B. Neodymium

This paragraph provides a review of the properties of neodymium lasers in GSGG host material. Singly doped and codoped crystals and their optical characteristics are compared to those of Cr:GSGG and Nd:YAG. The performance of Nd lasers in GSGG is compared to that in YAG.

According to Krupke et al [2, p. 102], Nd<sup>3+</sup>:GSGG was first demonstrated as a laser in 1976 by A. A. Kaminskii and his associates. This work must have been a little too late to have been covered by Koechner [1] in his extensive survey of solid-state laser materials. Little reference is made to GSGG as a host for any laser until 1982 and later. In recent years, however, intensive research has been conducted in Nd:GSGG lasers, particularly when codoped with chromium, as a new pulsed source with improved efficiency.

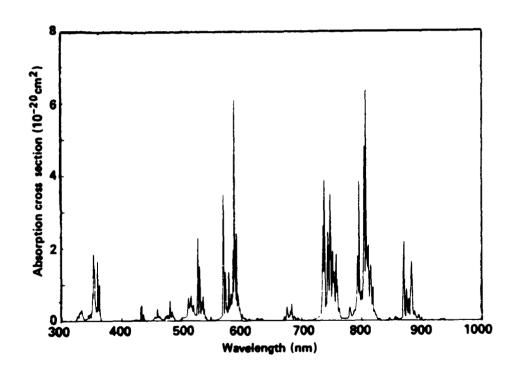
## 1. Singly Doped Nd:GSGG

If GSGG crystals are doped only with  $Nd^{3+}$ , a somewhat complex absorption spectra results. Figures 2 and 3 are examples as reported by researchers in Russia and the United States. The absorption spectra is of very fine structure with no broad bands as seen in the spectra for  $Cr^{3+}$  in Figure 1. Figure 3 is quite similar to the absorption spectra of Nd in YAG that is illustrated by Koechner [1, p. 57]. The result is a crystal that has a pale, pinkish-purple color like that of Nd:YAG. Most of the absorption occurs in the 700-900 nm region, which can be matched somewhat effectively (but not very efficiently) with xenon and krypton filled flashlamps.



Source: From page 97 of Reference 18.

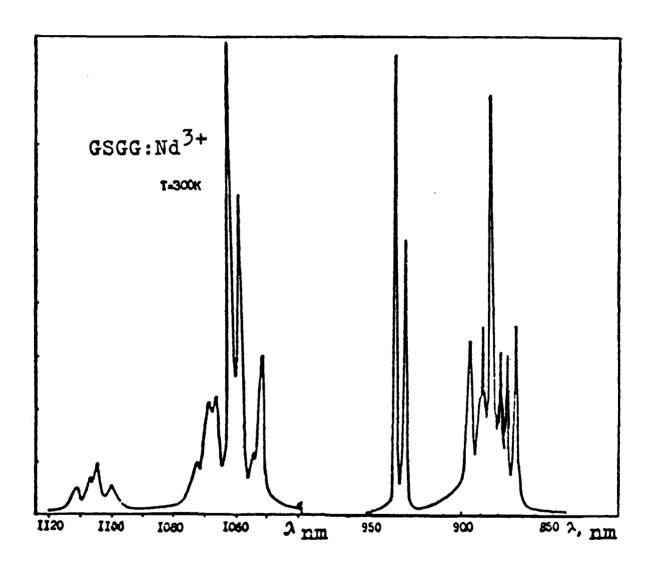
Figure 2. Nd3+ absorption spectra in GSGG at room temperature.



Source: From page 103 of Reference 2.

Figure 3. Spectral absorption cross section of Nd 3+ in GSGG at 296 °K.

Figure 4 is the fluorescence spectra of Nd<sup>3+</sup> in GSGG at room temperature. Denisov et al [18, p. 97] notes that the addition of chromium ions up to heavy doping concentrations does not change this fluorescence line shape. In Figure 4 there are several spectral lines besides the most familiar one at 1.06  $\mu$ m. These represent different radiative transitions that are possible in Nd<sup>3+</sup>.



Source: From page 98 of Reference 18.

Figure 4. Fluorescence spectra of Nd3+ in GSGG at room temperature.

The upper level of the lasing transition in Nd is the  $^4\mathrm{F}_{3/2}$  level. The terminal level for radiation at 1.06  $\mu m$  is the  $^4\mathrm{I}_{11/2}$  level. However, lower and higher energy transitions can also occur to the  $^4\mathrm{I}_{13/2}$  and  $^4\mathrm{I}_{9/2}$  levels, respectively. Figures 5, 6, and 7 (Source: From page 104 of Reference 2) show the normalized spectral emission cross section for each of these three transitions. The lines are not discrete, because the upper and lower transition levels are each split into two or more sublevel branches [1, pp. 55 and 56].

Since Figures 5, 6, and 7 have been normalized to unity, it is not obvious that the  $^4\text{F}_{3/2}$  to  $^4\text{F}_{11/2}$  transition (Fig. 6) dominates, at a wavelength of 1.06  $\mu$ m. However, Figure 4 does show, by a small margin, this to be the strongest emission line.

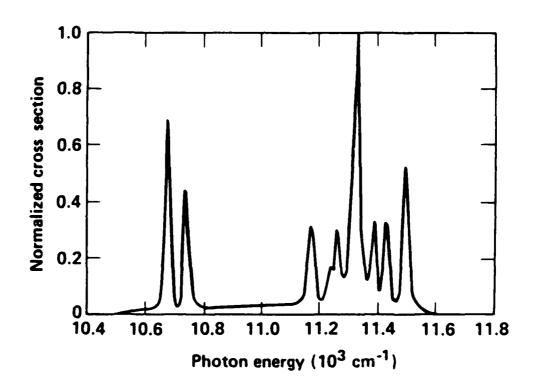
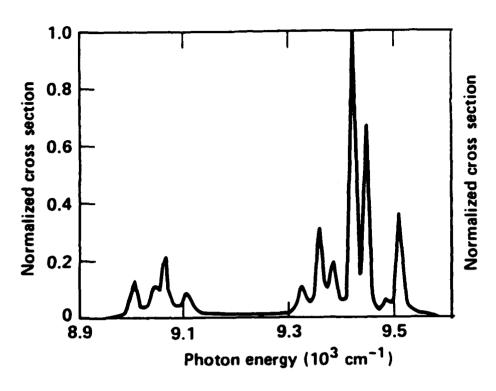


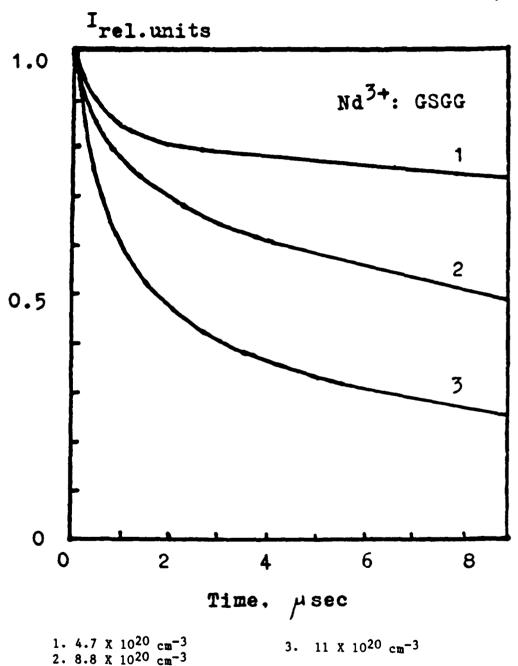
Figure 5.  ${}^{4}F_{3/2}$  to  ${}^{4}I_{9/2}$  transition.



1.0 0.8 0.6 0.4 0.2 0.8 7.0 7.2 7.4 7.6 Photon energy (10<sup>3</sup> cm<sup>-1</sup>)

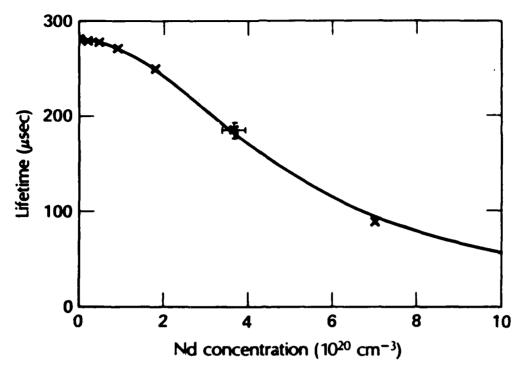
Figure 7.  $\frac{4F_{3/2} \text{ to } 4I_{13/2}}{4F_{3/2}}$ .

The fluorescence lifetime of Nd  $^{3+}$  in GSGG is about 270  $\mu s$ , for low concentrations, which is very close to that for Nd:YAG. If the Nd concentration is increased, there is a more rapid decay in the first few microseconds, followed by the conventional exponential decay thereafter. Figure 8 shows the decay curves for three neodymium concentrations. Figure 9 depicts the same effect in a different format. Both figures are for room temperature.



Source: From page 99 of Reference 18.

Figure 8. Nd3+ luminescence-decay curves in GSGG.



NOTE: Solid line is theoretical fit to data.

Source: From page 106 of Reference 2.

Figure 9. Concentration dependence of fluorescence lifetime of Nd<sup>3+</sup>
in GSGG.

## 2. Chromium Codoped Nd:GSGG

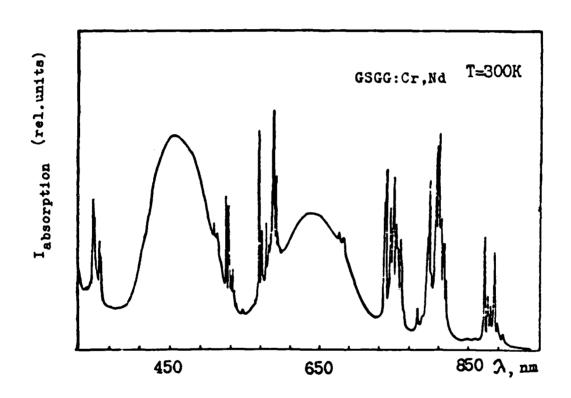
The broad band absorption spectra of  ${\rm Cr}^{3+}$  (Fig. 1) contrasted with the narrow absorption spectra of  ${\rm Nd}^{3+}$  (Figs. 2 and 3) leads to a natural conclusion that sensitizing a neodymium laser with chromium could improve pumping efficiencies, particularly for broad band flashlamp sources. This technique has proven useful in some laser hosts but not others.

During early investigations of Nd:YAG, Kiss and Duncan [19, p. 200] attempted to improve efficiency by doubly doping YAG with Nd and Cr. With the addition of Cr the Nd:YAG laser had a characteristic bright green color. Unfortunately, the transfer time of energy from Cr to Nd was found to be on the order of several milliseconds, which was much too long to benefit pulsed laser systems. However, it was hoped that cw operation, using broad band lamp pump sources, would be improved. For some configurations Kiss and Duncan showed a modest improvement, but their technique was not pursued.

The situation is quite different for Cr,Nd:GSGG, however, and sensitizing has proven so successful that little work is still ongoing in singly doped Nd:GSGG. As in YAG, codoping results in a brilliant green color [20, p. 36].

Codoping of Nd:GSGG with Cr was first demonstrated by Beimowski et al [8, pp. 234 and 235] and Pruss et al [9, pp. 355-358]. Efficient lasing at the nominal 1.06 µm line was obtained in a cw laser, using a krypton ion laser as a pump source. Within the same year Zharikov et al [21, pp. 1652-1653] demonstrated pulsed operation (normal mode and Q-switched) with a flash-lamp pump. In recent years, research conducted by a number of sources has continued to improve the performance of Cr,Nd:GSGG.

The absorption spectrum at room temperature of Cr,Nd:GSGG is shown in Figure 10. The Cr $^{3+}$  and Nd $^{3+}$  concentrations were 1 X  $^{10^{20}}$  and 2 X  $^{10^{20}}$  ions per cm $^3$ , respectively. The broad absorption bands of Cr $^{3+}$  (as in Fig. 1) are superimposed on the narrow Nd $^{3+}$  transitions (as in Figs. 2 and 3). This absorption spectrum is well balanced to the emission from xenon and krypton flashlamp pump sources, with a resulting increase in pump efficiency.



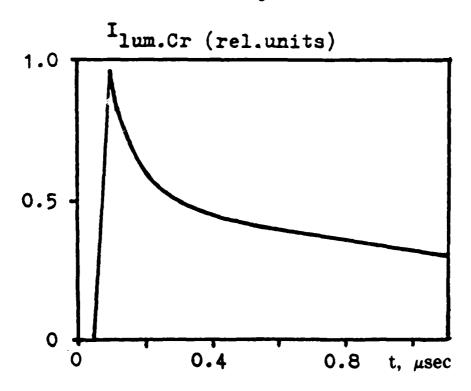
Source: From page 82 of Reference 22.

Figure 10. Absorption spectra of Cr3+ and Nd3+ in GSGG.

The excited  $Cr^{3+}$  donor ions, pumped by the broad absorption bands, transfer energy to the Nd $^{3+}$  acceptor ions, raising the Nd ions to the  $^{4}F_{3/2}$  upper transition level for lasing. The rate of energy transfer from Cr to Nd is faster than the decay rate of the excited Nd state. The population inversion in Nd is therefore enhanced by the additional pumping provided by the Cr ions. This process is shown in Figures 11 and 12. If a Cr,Nd:GSGG crystal is pumped in a chromium absorption band, there is a fast rise in excited Cr ions (Fig. 11), followed by a decay as energy is transferred to Nd ions. There is a corresponding rise in the number of excited Nd ions (Fig. 12).

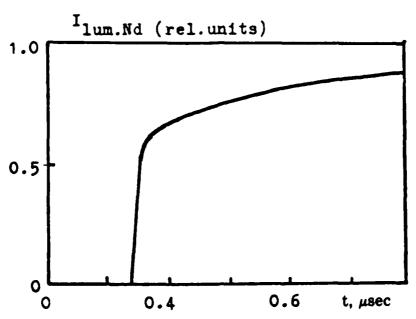
Figure 13 shows this process in another way. Here the decay curve of  ${\rm Cr}^{3+}$  luminescence in GSGG is shown with and without the presence of  ${\rm Nd}^{3+}$ . With the Nd acceptor ions, the excited states of Cr are depleted much more quickly.

Figure 14 shows the enhancement in the population of the  $^4\mathrm{F}_{3/2}$  excited state of Nd<sup>3+</sup> in GSGG and YAG, due to energy transfer from Cr<sup>3+</sup>. An instantaneous, step-like pulse excitation has been assumed. (The curves would be shifted to the right by about 100  $\mu s$  if a flashlamp pump had been assumed). Note that the curves for GSGG are much higher than those for YAG.



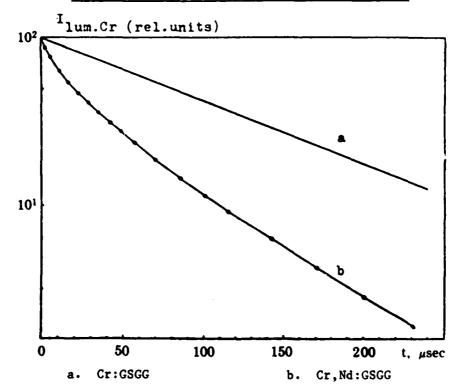
Source: From page 86 of Reference 22.

Figure 11. The excited-state decay curve of Cr3+ in Cr, Nd:GSGG.



Source: From page 87 of Reference 22.

Figure 12. The evolution of population of the upper Nd lasing level in Nd, Cr: GSGG, due to energy transfer from Cr.



Source: From page 88 of Reference 22.

Figure 13. Decay curves of Cr3+ luminescence.

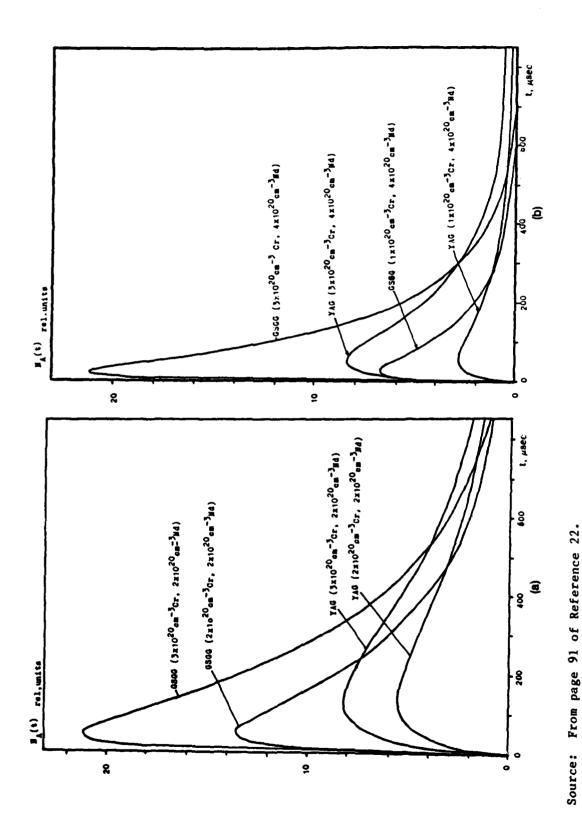
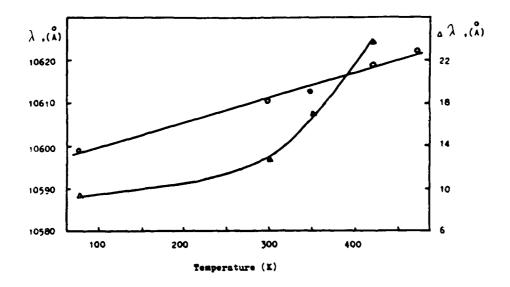


Figure 14. Enhancement of the excited state of Nd due to energy transfer from Cr in YAG and GSGG.

The spectral line on which lasing is achieved will vary slightly with temperature, as will the width of the fluorescence line. Figure 15 shows this temperature dependence as measured in one series of experiments.



Source: From page 99 of Reference 18.

Figure 15. Temperature dependence of the lasing line (o) and FWHM ( $\Delta$ ) of the line in Cr,Nd:GSGG.

#### V. CONCLUSIONS AND RECOMMENDATIONS

After reviewing the characteristics of Nd:GSGG and Cr,Nd:GSGG in comparison to Nd:YAG, the obvious question is: "Can we replace Nd:YAG with Cr,Nd:GSGG to provide better efficiency and a solution to all the problems identified in Section III.A?." Unfortunately the answer is "no" at this time, but "maybe" in the future.

For several years all reports on Cr,Nd:GSGG were positive. Any limitations were attributed to non-optimized crystal growth, stoichiometry, experimental laser components, etc. In 1983 GSGG laser efficiencies were being reported as 2.4 times higher than for YAG [23, p. 1306]. Statements were being made that "the presence of Cr3+ ions causes no deterioration in the optical quality of GSGG crystals" [24, p. 84]. Yet by 1985 it was evident that GSGG could not easily replace YAG in lasers for which beam quality and other factors are more important than efficiency.

Reed [25, p. 1625] reported on a series of laser experiments which have verified some of the problems with thermal distortion in GSGG. In addition, other drawbacks to the use of GSGG have been discovered. Following is a list of the disadvantages that have been identified with Cr,Nd:GSGG.

- Due to the broad pump bands in GSGG, a much higher thermal loading occurs. In addition, the thermal conductivity is only about two-thirds that of YAG. The result is thermal focusing in the laser rod which is six times worse than in YAG [25, pp. 1625-1627; 26, p. 320].
- The thermally induced birefringence in GSGG is much worse than in YAG. At high input levels, the output beam has a "spider-web" appearance and special Q-switch configurations must be used which can handle both polarization components [25, p. 1627; 26, pp. 317-320; and 27, pp. 106-108].
- Even for some single-pulsed laser applications as rangefinding, poor beam quality of GSGG has been a problem.
- The high cost and present availability of scandium as a raw material for crystal growth may be a problem [10, p. 79].
- The rods are opaque to HeNe and many other visible laser wavelengths, so optical alignment of resonator components is difficult.
- High crystal purities may be needed [28, pp. 283 and 284] to help overcome thermal problems.

All of these limitations do not mean that GSGG cannot be used. However, more complex optical schemes may be necessary. Potential solutions include oscillator-amplifier configurations [29, p. 1450], phase conjugation techniques, laser diode pumping, slab laser geometry, and other technologies now being addressed.

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